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Polarized Light Drives Anisotropic Expansion in Chalcogenide Glasses

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Common oxide glasses are transparent solids known for their inertness and stability. By contrast, their chalcogenide analogs (i.e. sulfides, selenides and tellurides) exhibit unexpected sensitivity to bandgap light, leading to several novel photoinduced effects. Particularly fascinating are the anisotropic effects created in their random structure by the polarized light. The in situ EXAFS experiments at NSLS provide the first atomistic evidence of the origin of such structural changes where the local structure around Se in an arsenic selenide glass film expands anisotropically depending on the polarization of the laser beam.

The exposure of solids to light leads to a number of important chemical reactions and physical phenomena in nature as well as in modern technology, e.g. photography, xerography, holography, photopolymerization, photosynthesis of plants, etc. In general, light interacts with a solid through its electronic states, which depend upon the constituent atoms and structure of the material. Light-induced change in the atomic structure involving displacement of atoms is unusual, except in special cases of *disordered* materials, including polymers and glasses. Such changes, when they occur, are expected to depend on the intensity and wavelength (l) of light. However, recently light-induced mass transport, photocrystallization, and an opto-mechanical effect have been reported on glassy chalcogenide films, which imply a change in the atomic structure that depends also on the *polarization* of light. Atomistic, chemical specific, experimental evidence about such changes has been missing. These so-called vector effects (because they depend on the direction of the electric field vector of light) are particularly intriguing because the starting structure of the unilluminated films is amorphous and isotropic.

To obtain atomistic insight into the light-induced structural changes that cause the newly discovered vector effects in chalcogenide glasses, it is necessary to have an experimental technique that is capable of probing the element-specific structure of amorphous materials in different directions as defined by the polarized laser beam. The extended x-ray absorption fine structure (EXAFS) analysis, which is conducted with a synchrotron light source, satisfies these requirements. Synchrotron light sources provide not only high-intensity but also *polarized* X-rays, which are essential for detecting anisotropic structures.

In the present work, the Lehigh University researchers, in collaboration with colleagues from Ohio, Pardubice (Czech Republic), and Cambridge (UK) universities and BNL, have used EXAFS analysis with polarized X-rays (in which the electrical field vector $\mathbf{E}_X$ // the ground plane) to probe the changes in the structure around Se and As atoms, and track the laser-polarization dependence of these changes in arsenic selenide glasses. The experiments were performed at the NSLS Beamline X18B; the schematic for *in situ* observations is shown in **Figure 1**. A Helium-Neon laser beam (λ=633 nm) intercepts the X-ray beam on the sample. A half wave plate is used to change the polarization direction of the laser beam (identified by its electric field vector, $\mathbf{E}_L$) so that

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$\mathbf{E}_L$ // $\mathbf{E}_X$ or $\mathbf{E}_L \perp \mathbf{E}_X$. Two different spots of the same sample were separately illuminated by two orthogonal, linearly polarized laser beams, and the structural changes were probed *in situ* by the X-ray beam.

Figure 2 shows an example of the light-induced structural changes for the two polarization directions of the laser beam. There is an expansion of the nearest-neighbor distance around the Se atoms - its magnitude depends on the direction of light polarization with larger expansion being observed when $\mathbf{E}_L$ // $\mathbf{E}_X$ than when $\mathbf{E}_L \perp \mathbf{E}_X$! Thus a vector structural change is observed *directly* for the first time for any amorphous material. The authors believe that the anisotropic local expansion around Se atoms occurs as the bandgap laser light excites and reorients the Se 4p lone pairs, and helps form anisotropic As-Se bonds. Their experiments provide an atomistic view of photo-induced vector phenomena in chalcogenide glasses, and guide the discovery of new materials with enhanced light-induced effects for photonic applications.

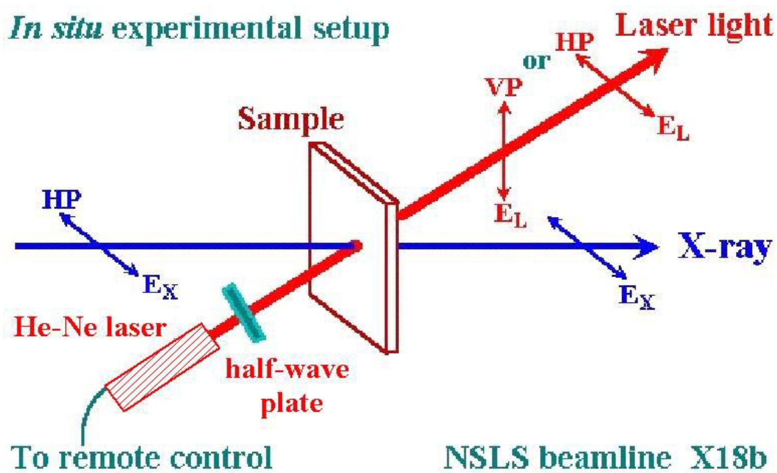

Figure 1. *In situ* EXAFS setup for studying laser-polarization-dependent photostructural changes. The a-As_2Se_3 film is irradiated with light of approximately the bandgap energy from a He-Ne laser (λ= 633 nm) and simultaneously observed by X-rays from the synchrotron. A half wave plate controls the polarization direction of the laser beam. HP: horizontal polarization. VP: vertical polarization.

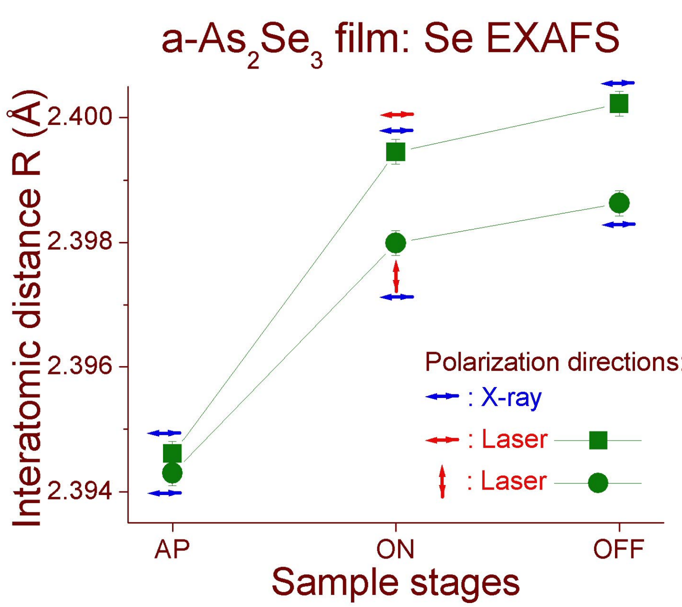

Figure 2: Effect of *in situ* laser irradiation on the interatomic distance from Se in amorphous As_2Se_3 film. Note a greater photoexpansion when $\mathbf{E}_L$ // $\mathbf{E}_X$ than when $\mathbf{E}_L \perp \mathbf{E}_X$. AP: as prepared; ON: laser beam is turned on; OFF: laser beam is switched off.